Spin dynamics of strongly doped $La_{1-x}Sr_xMnO_3$

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Cold neutron triple-axis measurements have been used to investigate the nature of the long-wavelength spin dynamics in strongly doped $La_{1-x}Sr_xMnO_3$ single crystals with $x=0.2$ and 0.3. Both systems behave like isotropic ferromagnets at low T, with a gapless ($E_0<0.02$ meV) quadratic dispersion relation $E=E_0+Dq^2$. The values of the spin-wave stiffness constant D are large ($D_{T=0}=167$ meV Å^2 for $x=0.2$ and $D_{T=0}=176$ meV Å^2 for $x=0.3$), which directly shows that the electron transfer energy for the d band is large. D exhibits a power law behavior as a function of temperature, and appears to collapse as $T\rightarrow T_C$. Nevertheless, an anomalously strong quasielastic central component develops and dominates the fluctuation spectrum as $T\rightarrow T_C$. Bragg scattering indicates that the magnetization near T_C exhibits power law behavior, with $\beta\simeq 0.30$ for both systems, as expected for a three-dimensional ferromagnet. © *1998 American Institute of Physics.* [S0021-8979(98)27311-7]

Since the recent discovery of unusually large magnetoresistive effects in perovskite manganites, the doped $LaMnO_3$ class of materials[1] has generated continued interest and has motivated experimental and theoretical work devoted to understanding of the origin of this colossal magnetoresistance (CMR) phenomenon. The large variation in the carrier mobility originates from an insulator-metal transition that is closely associated with the magnetic ordering. The on-site exchange interaction between the spins on the manganese ions is believed to be strong enough to completely polarize the (e_g) conduction electrons in the ground state, forming a ''half-metallic'' ferromagnet. However, hopping, and hence conduction, may only occur if the Mn core spins (formed by the d electrons in a t_{2g} orbital) on adjacent sites are parallel, which then directly couples ferromagnetic order with the electrical conductivity at elevated temperatures. This mechanism, known as the double exchange mechanism,[2] was first proposed in the 1950s, and has provided a good description of the evolution of the magnetic properties with band filling. However, in order to fully explain all the properties of the CMR materials, strong electron correlations,[3] and/or a strong electron-lattice coupling[4] in different polaronic approaches are invoked. This unique class of half-metallic ferromagnets provides an excellent opportunity to elucidate the influence of such correlations on the lattice and spin dynamics, which can best be probed by inelastic neutron scattering.

In the optimally doped regime with $x\sim 0.3$ it has been shown that the ground state spin dynamics is essentially that expected for a conventional metallic ferromagnet described by an isotropic Heisenberg model.[5–7] For the Ca-doped system, however, results obtained on polycrystalline samples[8] have indicated a possible coexistence of spin-wave excitations and spin diffusion in the ferromagnetic phase. In particular, it was suggested that it is this spin diffusion that drives the ferromagnetic phase transition rather than the thermal population of conventional spin waves. In the present publication we report diffraction and inelastic measurements of the spin dynamics in the metallic ferromagnets $La_{0.8}Sr_{0.2}MnO_3$ and $La_{0.7}Sr_{0.3}MnO_3$.

The single crystals used in the present neutron scattering experiments were grown at the Steel and Alloys Institute in Moscow, using the floating zone method. The crystals weighed 2.25 and 4.25 g, respectively. The samples were oriented such that the [100] and [010] axes of the rhombohedral $R\bar{3}c$ cell lie in the scattering plane. The neutron scattering measurements have been carried out on the NG-5 (SPINS) cold neutron triple-axis spectrometer at the NIST research reactor. The (002) reflection of pyrolytic graphite (PG) was used as monochromator and analyser for measuring the low-energy part of the spin-wave spectrum. We have used a flat analyzer with a fixed final energy $E_f=3.7$ meV, a cold Be filter on the incident beam, and collimations $40'$-S-$40'$-$130'$ in sequence from the neutron guide to detector. This configuration offered an energy resolution of ~ 0.15 meV, together with good q resolution. Each sample was placed in a helium-filled aluminum cell in a displex refrigerator. The sample temperature ranged from 15 to 325 K for $La_{0.8}Sr_{0.2}MnO_3$, and from 30 to 375 K for $La_{0.7}Sr_{0.3}MnO_3$, and was controlled to within 0.1°.

The crystal structure of both systems at room temperature and below is rhombohedral ($R\bar{3}c$), with $a_0\simeq b_0\simeq c_0\simeq 3.892$ Å for $x=0.2$ and $a_0\simeq b_0\simeq c_0\simeq 3.884$ Å for $x=0.3$.

Figure 1 shows the integrated intensity of the (100) Bragg reflection as a function of temperature for both samples. This reflection has a finite nuclear structure factor, and therefore the intensity in the paramagnetic phase is nonzero. The increase in intensity below T_C is due to magnetic scattering produced by the ferromagnetism of spins aligning

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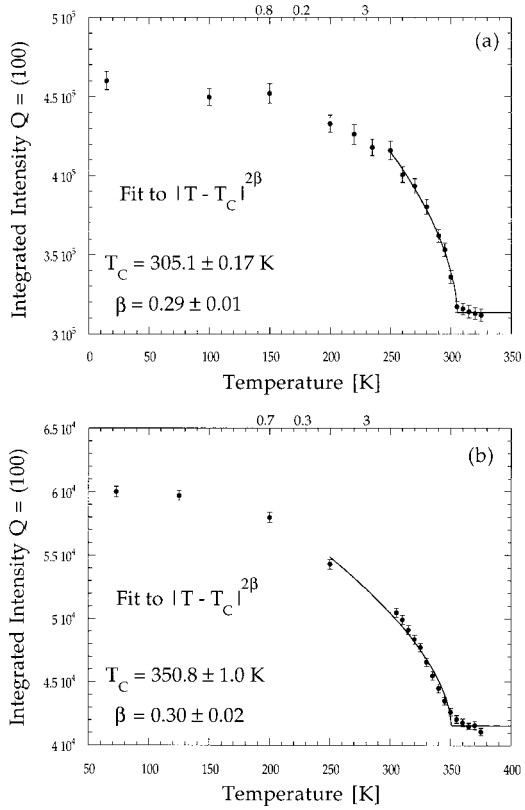

FIG. 1. Temperature dependence of the integrated intensity of the (100) Bragg peak for (a) $La_{0.8}Sr_{0.2}MnO_3$ and (b) $La_{0.7}Sr_{0.3}MnO_3$. There is a nuclear contribution to this peak, and the additional temperature-dependent intensity originates from the onset of the ferromagnetic order at $T_C = 305$ K for the $x=0.2$ system, and $T_C = 350.8$ K for $x=0.3$. The solid curves are fits of the points near T_C to a power law.

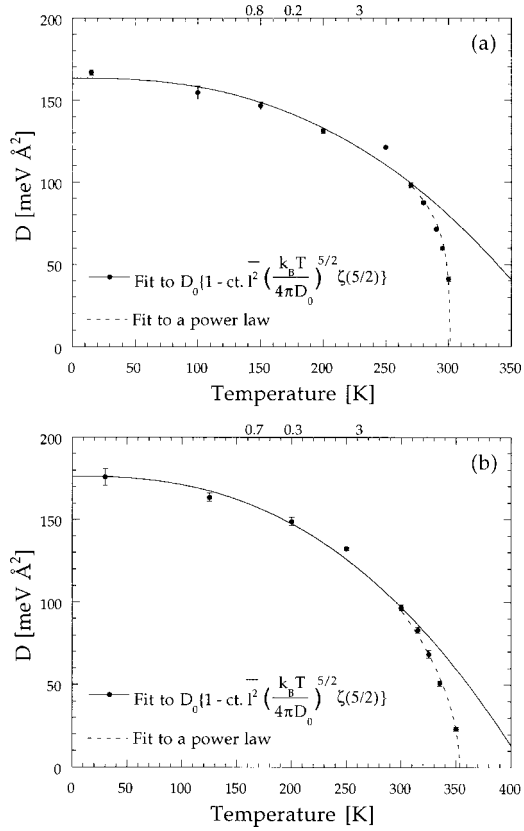

FIG. 2. Spin-wave stiffness coefficient D in $E=E_0+Dq^2$ as a function of temperature for (a) $La_{0.8}Sr_{0.2}MnO_3$ and (b) $La_{0.7}Sr_{0.3}MnO_3$. The solid curves are fits to Eq. (1). D appears to vanish at the ferromagnetic transition temperature, as expected for a conventional ferromagnet. The dashed curves are fits to a power law.

on the manganese ions and yielding a magnetic structure factor. The solid curve is a fit of the points near T_C to a power law. The best fits give $T_C = 305.1$ K and a critical exponent $\beta = 0.29 \pm 0.01$ for $La_{0.8}Sr_{0.2}MnO_3$, and $T_C = 350.8$ K and $\beta = 0.30 \pm 0.02$ for $La_{0.7}Sr_{0.3}MnO_3$. Both values of the critical exponent are slightly below, but rather close to, the well known three-dimensional Heisenberg ferromagnet model value of $\sim 1/3$.

We have investigated the spin dynamics in the (1,0,0) high-symmetry direction in both samples. The ground state spin dynamics for a half-metallic ferromagnet was not expected to differ much from the conventional picture of well defined spin waves, and we found that the long-wavelength magnetic excitations were in fact the usual spin waves, with a dispersion relation given by $E=E_0+Dq^2$, where E_0 represents the spin-wave energy gap and the spin stiffness coefficient is directly related to the exchange interactions. The spin-wave gap E_0 was too small to be measured directly in energy scans at the zone center, but very high-resolution measurements on the NG-5 (SPINS) cold-neutron triple-axis spectrometer have allowed us to determine that $E_0 < 0.02$ meV for both systems, which demonstrates that these are ''soft'' isotropic ferromagnets. A previously reported value of $E_0 = 0.75$ meV for the $x=0.3$ system[6] was obtained from an extrapolation of higher q data, not from direct high-resolution measurements as in the present case. The low-temperature values of the spin-wave stiffness constant D are large: $D_{T=0} = (166.8 \pm 1.51)$ meV Å^2 for $x=0.2$ and $D_{T=0} = (176 \pm 5.00)$ meV Å^2 for $x=0.3$, and give a ratio $D/k_B T_C \sim 6.34$ Å^2 and 5.82 Å^2 for the $x=0.2$ and 0.3 systems, respectively. Both values are quite large, as might be expected for an itinerant electron system.

Figure 2 plots the temperature dependence of the spin-wave stiffness D. The data have been analyzed in terms of two-spin-wave interactions in a Heisenberg ferromagnet within the Dyson formalism,[10] which predicts that the dynamical interaction between the spin waves gives, to leading order, a temperature dependence:

$$D(T) = D_0 \left\{ 1 - \frac{v_0 \overline{l^2} \pi}{S} \left(\frac{k_B T}{4\pi D_0} \right)^{5/2} \zeta\left(\frac{5}{2}\right) \right\}, \tag{1}$$

where v_0 is the volume of the unit cell, S is the average value of the manganese spin, and $\zeta(5/2)$ is the Riemann zeta integral. $\overline{l^2}$ is the moment defined by $\overline{l^n} = S/3D\{\Sigma l^{n+2} J(\mathbf{1})\}$ and which, compared to the square of the lattice parameter a^2, gives information about the range of the exchange interaction. The solid curves in Fig. 2 are fits to Eq. (1), and are in good agreement with the experimental data for reduced temperatures $t = (T - T_C)/T_C$ up to $t_1 \simeq -0.1$ for $La_{0.8}Sr_{0.2}MnO_3$ and -0.14 for $La_{0.7}Sr_{0.3}MnO_3$. The fitted values of $\overline{l^2}$ give $\sqrt{\overline{l^2}} = (3.92 \pm 1.04) a_0$ for $x=0.2$, and $\sqrt{\overline{l^2}} = (3.84 \pm 1.22) a_0$ for $x=0.3$, which indicates that the exchange interaction extends beyond nearest neighbors in both

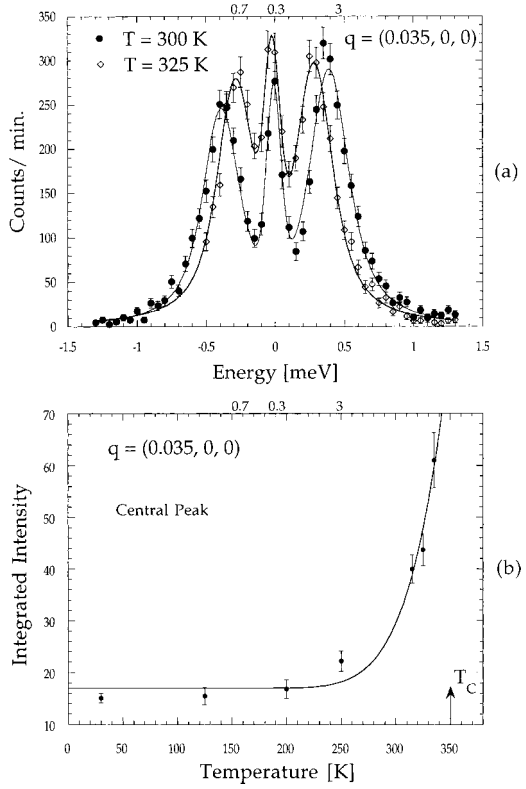

FIG. 3. (a) Constant-q magnetic inelastic spectra collected at 300 and 325 K and a reduced wave vector vector $q=(0,0,0.035)$ for $La_{0.7}Sr_{0.3}MnO_3$ ($T_C=350.8$ K), and (b) temperature dependence of the integrated intensity of the quasielastic central component. The dominant effect is the development of a strong quasielastic component in the spectrum. Above T_C, all the scattering in this range of q is quasielastic.

systems. For $t>t_1$, the experimentally measured values of D depart from the $T^{5/2}$ dependence, having rather a power law behavior and appearing to collapse as $T\rightarrow T_c$. The dashed lines in Fig. 2 are fits to a power law $[1-(T/T_C)]^{\nu'-\beta}$, where ν' is the critical exponent for a three-dimensional ferromagnet.

In the course of our measurements we have noticed that the central peak has a strong temperature dependence on approaching T_C, while typically the central peak originates from weak temperature-independent nuclear incoherent scattering. Figure 3(a) shows two magnetic inelastic spectra collected at 300 and 325 K, and reduced wave vector $q=0.035$ away from the (100) reciprocal point in $La_{0.7}Sr_{0.3}MnO_3$ ($T_C=351$ K). A flat background of 4.9 counts plus an elastic incoherent nuclear peak of 110 counts, measured at 30 K, have been subtracted from these data. We can clearly see the development of the quasielastic component, comparable in intensity to the spin waves, and the temperature dependence of the strength of this scattering is shown in Fig. 3(b) as a function of temperature. We observe a significant intensity starting at 250 K (~100 K below T_C), and the scattering peaks at T_C. At and above T_C all the scattering is quasielastic. For typical isotropic ferromagnets, such as Ni, Co, Fe, any quasielastic scattering below T_C is too weak and broad to be observed directly in the data, and can only be distinguished by the use of polarized neutron techniques. In Fig. 3(a) we can nevertheless see that the spectrum starts to be dominated by this quasielastic component at temperatures well below T_C. The appearance in the ferromagnetic phase of a quasielastic component was first observed on Ca-doped polycrystalline samples,[8] and it has been suggested that it is associated with the localization of the e_g electrons on the Mn^{3+}/Mn^{4+} lattice, and may be related to the formation of spin polarons in the system.[9] We have observed a similar anomalous behavior of the central peak in the more lightly doped system $La_{0.85}Sr_{0.15}MnO_3$,[11] but for that doping we find that the central component becomes evident only much closer (~25 K) to the Curie temperature. It thus appears that the coexistence of spin-wave excitations and spin diffusion is a common characteristic for many perovskite manganites, and that it may be relevant for the giant magnetoresistance property of these systems. It is therefore important to pursue the study of this aspect with polarized neutron techniques, in order to determine the nature of the fluctuations involved in this new quasielastic component to the fluctuation spectrum.

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